Comparison of Empirically Derived Ozone Losses in the Arctic Vortex

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Abstract

A number of studies have reported empirical estimates of ozone loss in the Arctic vortex. They have used satellite and *in situ* measurements and have principally covered the Arctic winters in the 1990s. While there is qualitative consistency between the patterns of ozone loss, a quantitative comparison of the published values shows apparent disagreements. In this paper we examine these disagreements in more detail. We choose to concentrate on the five main techniques (Match, SAOZ/REPROBUS, MLS, vortex average descent and the HALOE ozone-tracer approach). Estimates of the ozone losses in three winters (1994/95, 1995/96 and 1996/97) are re-calculated so that the same time periods, altitude ranges and definitions of the Arctic vortex are used. This recalculation reveals a remarkably good agreement between the various estimates. For example, a superficial comparison of results from Match and from MLS indicates a big discrepancy (2.0 ± 0.3 ppmv and 0.85 ppmv respectively, for air ending at ~460K in March 1995). However the more precise comparisons presented here reveal good agreement for the individual MLS periods (0.5 ± 0.1 vs 0.5 ppmv; 0.4 ± 0.2 vs 0.3-0.4 ppmv; and 0.16 ± 0.09 ppmv vs no signficant loss). Initial comparisons of the column losses derived for 1999/2000 also show good agreement with four techniques giving 105 DU (SAOZ/REPROBUS), 80 DU (380-700K partial column from POAM/REPROBUS), 85 ± 10 DU (HALOE ozone-tracer) and 88 ± 13 (400-580 partial column from Match). There are some remaining discrepancies with ozone losses calculated using HALOE ozone-tracer relations: it is important to ensure that the initial relation is truly representative of the vortex prior to the period of ozone loss.

Introduction

Long-term decreases in total ozone have been observed at high and mid-latitudes of both hemispheres [e.g., *Staehelin et al.*, 2001, and references therein]. The most dramatic losses are observed over Antarctica during each Austral spring – in recent years, over 99% of the ozone is removed from altitudes between 15 and 19 km during a period from mid August to October. Total ozone measured at Halley Bay, Antarctica (69˚S) in October averaged 160 Dobson Units in the 1990s, 40% that of the long-term average [*Jones and Shanklin*, 1995].

The information about the long-term evolution of the ozone layer over the Arctic is less good than over the Antarctic, a situation brought about by the lack of long-term measurements within the Arctic circle (there is very little land mass there) and the larger interannual and intraseasonal variability. However unusually low total ozone was observed in several Arctic winters in the 1990s. For instance, low values of total ozone (up to 45% below the 1957-1992 average) were observed over the Canadian High Arctic (north of 70°N) in March 1996 and 1997 [*Fioletov et al.*, 1997]. However, such low values are not seen in all Arctic winters, as is now the case in the Antarctic, and unusually low values were also observed in winters (e.g., 1967) during which chemical effects are expected to have been negligible [*Fioletov et al.*, 1997]. It is thus apparent that low ozone amounts in the Arctic result from both chemical and dynamic factors which vary greatly from winter to winter, and whose magnitude and relative importance are hard to quantify.

The first quantitative estimates of widespread chemical ozone loss in the Arctic lower stratospheric vortex were for the 1988/89 winter [*Proffitt et al.*, 1990; *Schoeberl et al.*, 1990]. Since then a number of studies have evaluated the chemical ozone loss that has occurred in particular winters with increasing emphasis on its quantification. For the sake of clarity, we use the term ‘ozone loss’ to mean a chemical change that has occurred in defined air masses over a period of days or weeks, and the term ‘ozone trend’ to describe

the long-term changes that have been observed over recent decades – of whatever cause. These two terms are frequently used interchangeably and it seems useful to draw a distinction. The results of studies based primarily on measurements that have produced empirical estimates of the ozone loss have been quite diverse. It is hard to compare them. In general (though not always) it seems that the empirical methods produce higher ozone loss rates in the Arctic vortex than do the photochemical models [e.g., *Becker et al.*, 1998, 2000; *Guirlet et al.*, 2000; *Kilbane-Dawe et al.*, 2001; *Sinnhuber et al.*, 2000]. Given this discrepancy, it is important to know how well the various empirical approaches agree.

Arctic vortex

The main feature of the Arctic winter stratosphere is the Arctic vortex, which, in conjunction with the Aleutian high, dominates the high latitude circulation from November to March [e.g., *Schoeberl et al.*, 1992; *Manney et al.*, 1995a]. The vortex forms in autumn as the stratosphere cools radiatively. Air moving poleward is acted on by the Coriolis force and an increasingly strong westerly jet stream develops. The vortex extends from the lower stratosphere to the mesosphere. The average temperatures inside the vortex are significantly lower than in the surrounding air, but on any given day the lowest temperatures can either be inside or straddle the edge of the vortex.

During each Arctic winter, the vortex is buffeted by a series of stratospheric sudden warmings associated with planetary-scale waves which originate in the troposphere. During these events the vortex is displaced off the pole, often by considerable amounts. In the case of major warmings, it may break down temporarily resulting in a brief period of polar zonal mean easterlies during winter. The final, springtime vortex breakdown in the Arctic is much earlier than in the Antarctic, and it is often precipitated by wintertime warmings. The area of the Arctic vortex, the strength of the sudden warmings and the timing of the final breakdown vary enormously from year to year.

The edge of the vortex can be defined using potential vorticity (PV). In ozone loss studies a PV value typical of the edge region is often used, as this is found in practice to give equivalent results to using the more general maximum gradient in PV. As is shown later, it is important to be careful in defining the vortex edge when comparing empirically derived ozone losses. PV is measured in PV units (1 PVU = 10^{-4} km^2 kg s^{-1}) and the vortex edge has to be defined by different PV values on different isentropic surfaces. To make comparison between different levels easier, the concepts of scaled and normalised PV are used in studies described here. Scaled PV (defined as PV/g(dϑ/dp), where dϑ/dp is a standard atmosphere value) is a relatively height-independent quantity. Normalised PV (defined as 2.65 x 10^5 scaled PV) is also height-independent and is additionally chosen to have the same numeric values (in s^{-1}) as Ertel's PV (in PVU) on the 475 K surface (see *Rex et al.* [1999] for more detailed discussion).

The wave activity causes the Arctic vortex to be noticeably warmer and more dynamically variable than its Antarctic counterpart. On average the vortex temperatures in the Arctic are 10-20 K higher than in the Antarctic [e.g., *Manney et al.*, 1996a]. There is a large variability in Arctic ozone on both short (day-to-day) and long (year-to-year) time-scales. This variability in ozone is caused by the variability in the transport of air in the stratosphere, in the tropospheric forcing, and by variations in the chemical ozone loss.

The chemical processes causing ozone loss in the Arctic lower stratosphere depend critically on temperature, and they are basically the same as those causing the ozone loss in the Antarctic (e.g., *Ravishankara and Shepherd* in WMO [1999] and references therein). At temperatures below about 190-195K, heterogeneous chemical reactions can occur on the surfaces of cold particles such as Polar Stratospheric Clouds (PSCs) that convert inactive inorganic chlorine-containing species such as $ClONO_2$ and HCl to active forms that can catalytically remove ozone. The amount of ozone destroyed during each activation episode is controlled by factors such as the size and location of the cold areas, the amount of

sunlight present to drive the ozone loss catalytic cycles, and the rate of the subsequent deactivation through reaction of ClO with NO_2 to form $ClONO_2$. Deactivation is slower in mid-winter when ambient NO_2 concentrations are low (because the photolysis of HNO_3 and its reaction with OH are slow) or in air where HNO_3 has been permanently removed through sedimentation of particles ('denitrified').

The variability in the meteorology of the Arctic vortex has important chemical consequences. The area in which the temperature is below that at which PSCs can potentially form varies a great deal from year to year [*Pawson and Naujokat*, 1997, 1999]. Further, the timing of the cold periods, the position of the cold areas within the vortex, and the position of the vortex when they occur determine the amount of sunlight available to drive the chemical ozone loss and the volume of air processed through cold regions where chlorine can be activated. The combination of these factors means that the highly variable Arctic meteorology causes large year-to-year variability in the amount of chemical ozone loss. In general more chemical ozone loss is expected in colder winters than in warmer winters.

This variability underlies how important it is to know how much ozone there would have been if no chemical ozone loss had taken place. The problem is complicated by the fact that the magnitudes of the ozone changes resulting from the dynamic variability, the chemical ozone loss, and the underlying seasonal cycle are similar. Studies of ozone loss need to be able to separate these effects.

Methods of Estimating Ozone Loss

Empirical estimates of ozone loss in the Arctic vortex have been reported in a range of studies based on a few measurements at a single site in one year to many measurements made at many locations in several winters. In this section we describe the main types of approach that have been used to estimate chemical ozone loss, concentrating on the five techniques that have been used in several winters.

The movement of air masses is clearly the most important natural influence on the ozone fields at high northern latitudes. Measurement-based estimates of ozone loss must be able to distinguish between these motions and photochemical loss. The fastest air motions are horizontal and often reach 100 ms^{-1}. On time-scales of a few days, these horizontal motions occur on isentropic surfaces and are well captured in standard meteorological analyses. The slower motion is vertical: local vertical velocities can be 1 ms^{-1}, while the slow radiative cooling of the air in the vortex (1 K/day), the diabatic descent, is equivalent to a downward velocity of the order of 0.1 cm s^{-1}. These need to be taken into account in empirical calculations of ozone loss.

Broadly speaking the various approaches can be split into two categories:

1) studies where the effects of transport are calculated explicitly using transport calculations driven by winds, temperatures, etc., based on meteorological analyses; and
2) studies where the effects of transport are implicitly allowed for by using measurements of long-lived tracers.

These are now considered in turn.

Match

The Match technique has been developed to estimate chemical ozone loss rates in the lower stratosphere as directly as possible [*von der Gathen et al.*, 1995; *Rex et al.*, 1997, 1998, 1999, 2001; *Schulz et al.*, 2000, 2001]. Results from a similar approach using ozone measurements by the ILAS satellite instrument have been reported for the 1996/97 winter [*Sasano et al.*, 2000]. Match is a pseudo-Lagrangian technique based on the identification of air parcels whose ozone amount is measured twice within a ten day period. Any difference is ascribed to chemical loss. In practice a large number of such 'matches' are required to obtain statistically significant results. The identification of these air masses is

achieved by calculating 3D air mass trajectories with the horizontal (isentropic) component based on winds from ECMWF analyses and with the vertical (cross-isentropic) motions derived from the radiative cooling rates calculated by the SLIMCAT chemical transport model (CTM) [*Chipperfield*, 1999] using the MIDRAD radiation scheme [*Shine*, 1987] with ECMWF wind and temperature analyses, and climatological ozone values. During Match campaigns, forward trajectories are calculated from ozone sonde flights until that air mass can be sampled by a second ozone sonde (up to a limit of 10 days). The average length of a Match trajectory is 5-6 days. An air mass is considered to be successfully intercepted if the displacement between the end of a trajectory and the location of the second intercepting ozone sonde is less than about 500 km. By performing a large number of such matched measurements over a range of altitudes during a winter, the evolution of the vortex average ozone loss can be reconstructed with high vertical (10 K or better) and temporal (2 week) resolution.

The air masses inside the polar vortex can experience substantial strain and stirring. Match depends on identifying those air masses that are well conserved, entailing a careful selection procedure [*Rex et al.*, 1999]. The criteria used include rejecting ozone sonde profiles which show large changes over short vertical intervals; accepting limited dispersion within a cluster of 6 companion trajectories calculated above, below and to each side of the central trajectory; and limiting the PV variation along the trajectories. In order to determine whether a measurement takes place inside or outside the vortex, a normalised PV is calculated along the air mass trajectories. If it exceeds 36 PV units, the measurement is considered to have been inside the vortex.

In each winter, the Match trajectories are examined to check that the vortex is sampled homogeneously, so that the results reflect vortex averaged conditions. Ozone loss can occur preferentially in some regions inside the vortex depending on the radiation and temperature fields during a given winter. For example, in the 1996/97 winter the ozone loss rates were

greater towards the centre of the vortex due to greater occurrence of low temperatures in this region [*Schulz et al.*, 2000].

MLS

Manney et al. [1996b] analysed the ozone loss during some time periods in winter 1994/95 by using MLS measurements of ozone and trajectory calculations to account for transport effects. Horizontal winds from the UKMO data assimilation system and vertical velocities from the radiation code MIDRAD driven by UKMO temperatures are used for the trajectory calculations. Thus the MLS and Match approaches use the same radiation code, but with different temperatures driving it. Reverse trajectory calculations are started at all points on the gridded MLS data between 465 and 840K and run backward in time for a few weeks. Previous MLS measurements are interpolated to the starting points of these trajectories. This approach has been applied to measurements of long-lived tracers (N_2O and CH_4) and of ozone. Manney et al. [1995c] show that the advected tracer fields agree well with the observations, suggesting that the trajectory calculations are sufficiently precise. Hence, differences in ozone are attributed to chemistry. The differences in ozone are then averaged over the polar vortex, which is defined by the 1.3×10^{-4} s^{-1} isoline of scaled PV (= 34 s^{-1} normalized PV), so that the results represent the vortex averaged ozone loss, even if the ozone vmr or the ozone loss within the vortex is not homogenous. The same method has been used to infer chemical ozone losses for some periods during the 1993/94 [*Manney et al.*, 1995a], 1995/96 [*Manney et al.*, 1996a] and 1996/97 [*Manney et al.*, 1997] winters. Both this approach and one that is only slightly different (based on forward trajectories from a regular grid and interpolation of the MLS measurements onto this grid) were used for periods during the 1992/93 winter [*Manney et al.*, 1995b].

Vortex average

A similar conceptual approach based on ozonesonde measurements was used for early 1997 by *Knudsen et al.* [1998] and for 1991/92 by *Lucic et al.* [1999]. *Knudsen et al.* calculate

the trajectories on isentropic surfaces (350-675K) from ECMWF analyses. However rather than following the large number of individual trajectories, *Knudsen et al.* [1998] calculate the bulk vertical advection of the average ozone profile in the vortex from diabatic cooling rates calculated using the ECMWF operational heating scheme with ozone mapped in PV-theta space and a constant 5 ppmv H_2O. Transport of ozone across the vortex edge through mixing is calculated by tracking the trajectories in this region. The vortex edge is defined as the maximum in the PV gradient with respect to equivalent latitude, smoothed over 3 days.

Observed vs CTM-calculated 'passive' ozone

3D transport models driven by meteorological analyses can be used to simulate what would happen to ozone if no chemical loss took place (passive ozone). The difference between measurements and the simulated passive ozone can be ascribed to chemical ozone loss. The 3D circulation model used in many of these studies is the REPROBUS (Reactive Processes Ruling the Ozone Budget in the Stratosphere) CTM [*Lefèvre et al.*, 1994; *Lefèvre et al.*, 1998]. It extends from the ground up to 10 hPa, with a horizontal resolution of 2° latitude by 2° longitude. REPROBUS is driven by the 6-hourly ECMWF meteorological analyses. Vertical motions are calculated directly from the analysed ECMWF residual vertical velocity fields. It is initialised in December of each winter, the exact date depending on the formation of the vortex and the first cold period where activation could take place. The ozone field is initialised using MLS data, usually version 4 (as used in *Manney et al.* [1996b]). Comparisons with long-lived tracer measurements performed in the Arctic in 1995 [*Goutail et al.*, 1999] and 1997 [A. Engel, personal communication] have shown that despite the upper limit at 10 hPa, diabatic descent was reasonably well reproduced by the ECMWF analysis and the semi-lagrangian transport scheme of REPROBUS for a 3-4 month period.

This approach has been used by *Goutail et al.* [1999] using the high latitude measurements from the ground-based and balloon-borne SAOZ instruments which are inside the vortex at

475K (PV > 42 PVU) and by *Deniel et al.* [1998] using ozone profile measurements from POAM. The ozone losses derived from the two data sets are consistent. The principal diagnostic of ozone loss produced using this technique is the vortex-averaged, column ozone loss from the beginning of the integration, with the results usually presented as percentage column losses. Vertical profiles of the ozone loss are derived using the POAM and SAOZ balloon data. A similar approach has been applied for ozone profile measurements made at a single site [*Hansen et al.*, 1997]. This is comparable to vortex average studies as long as sufficient measurements are made inside the vortex as it moves around to ensure that it is well sampled, or that the vortex is homogeneous.

HALOE tracer relations

The final technique considered here removes the effects of transport implicitly by the use of tracers measured by HALOE [*Müller et al.*, 1996, 1997ab, 1999, 2001]. In a homogeneous air mass, a non-linear compact relation between ozone and an inert tracer will only change if there is mixing with other air masses with different compositions or if there is any chemical change in the ozone (see *Müller et al.* [2001] for full discussion). In the absence of mixing into the vortex, any change in the relation between ozone and an inert tracer inside the vortex can be ascribed to a chemical change in ozone. Initially this idea was used by *Proffitt et al.* [1990, 1993], and it has been developed by *Müller et al.* [1996, 1997a,b, 1999] using HALOE measurements of O_3, CH_4 and HF to estimate ozone loss in several winters. There is limited sampling of the vortex during early and mid winter as HALOE is a solar occultation instrument and UARS has two measurement modes and covers the northern high latitudes every other 35 days.

One potential confusion in the published results arises from the revision of the HALOE data from version 17 to version 18 that affected the results of the early analyses [*Müller et al.*, 1996]. A reanalysis of the derived ozone loss for the first four winters based on version 18 data yielded larger calculated ozone losses than for version 17 data [*Müller et al.*, 1997a].

The reason for this is a combination of several smaller changes in the HALOE data, which sum up to the observed effect. First, O_3 mixing ratios are higher in version 18. The increase is larger at higher altitudes and for larger O_3 mixing ratios, so that the estimated ozone loss increases. Further, CH_4 mixing ratios at lower altitudes (pressures greater than about 50-70 hPa) have decreased from versions 17 to 18, which also leads to an increase in the calculated ozone loss. Both effects are responsible for the fact that the ozone loss calculated previously from version 17 data was underestimated [*Müller et al.*, 1999]. The current version of HALOE data is version 19. Owing to the much smaller change of the HALOE O_3 and CH_4 data between versions 18 and 19 than between versions 17 and 18, the ozone losses derived from the version 19 data are likely to deviate only slightly from losses deduced from version 18 data. Preliminary analysis of the 1996/97 winter with version 19 data indicates that somewhat lower column losses will be derived than from version 18 data.

Comparisons of results

Comparisons of the results from the different approaches used to infer ozone loss in the Arctic are hampered by the fact that the altitude range, horizontal extent (vortex definition) and time periods used by the published works are different. These differences are partly unavoidable due to the constraints of the data sets used. However in many cases the different data sets can be reanalysed for certain time periods and regions where they overlap. Results from these reanalyses can be compared directly. Comparisons between pairs of techniques which explicitly allow for transport are presented first, and these are then compared to the tracer correlation technique where transport is implicitly accounted for.

Match - MLS

An example of how the relatively small differences in analysis criteria can affect the derived ozone losses is shown in Figure 1. For the winter 1994/95, *Manney et al.* [1996b] calculated the accumulated ozone losses in subsiding layers of air for three different time periods (heavy black lines in Figure 1). In *Rex et al.* [1999] the ozone loss rates from the

Match study were accumulated in several subsiding layers of air over one long time period (dashed thin black lines in Figure 1). At first glance a comparison of loss rates seems possible as one of the layers analysed in the original Match study (heavy blue line in Figure 1) is close to the three layers analysed by *Manney et al.* [1996b]. However, the vertical offset in the region of large vertical gradients in the ozone loss rate prevents a meaningful comparison of the results. Furthermore in the MLS analysis the vortex edge is defined 34 s^{-1} nPV (given as 1.2 x 10^{-4} s^{-1} scaled PV in the paper), whereas the 36 s^{-1} isoline of nPV is used in the Match study. If one were to ignore these facts and compare the techniques by combining the MLS results to cover one long time period, one would find that Match gives an accumulated loss of 2.0 ± 0.3 ppmv in that layer [*Rex et al.*, 1999], while the combined loss for the three MLS periods is 0.85 ppmv [*Manney et al.*, 1996b]. This superficial comparison indicates, wrongly, that there are considerable discrepancies between the published results from the two techniques.

Here we present results from a reanalysis of the Match data set using the same definition of the vortex edge, the same time periods and the same altitude regions which were used in *Manney et a*l. [1996b]. The results of the reanalysis of the Match data and the published results from MLS are given in Table 1 and shown in Figure 2. Three time periods are compared for the 1994/95 winter. For the latter two periods Match has been analysed for exactly the same periods of time as in the MLS study and the results agree within the experimental uncertainty. The overlap during the first period is not perfect: the starting date for the first period in the MLS study is dictated by the date when the MLS instrument turned south on 31 December 1994, while the Match results are available from 1 January 1995 on. However due to the lack of sunlight during mid-winter it is not very likely that a significant fraction of the accumulated ozone loss in the whole MLS period (21 December 1994 to 1 February 1995) occurred during the first nine days that are not included in the Match study. This supposition is consistent with very low ozone loss rates found by Match in early January, as shown in Figure 1. Overall the results from the MLS and the Match analyses

agree well for the 1994/95 winter.

A similar procedure has been performed to compare the Match and MLS results for the 1995/96 and 1996/97 winters, with the Match results being reanalysed to match the time periods, vertical regions and vortex edge definitions in which results are available from the MLS study. The results of these reanalyses are shown with the MLS results in Figure 2 and in Tables 2 and 3. The agreement for the 1995/96 winter when the MLS criteria could be used exactly is very good. However for 1996/97 exactly the same criteria could not be used. The time period used in the MLS study is from 26 February to 12 April 1997, with the end date dictated by the start of the north viewing period of MLS. The Match results are only available until 31 March 1997. Thus the last 12 days in the MLS study cannot be included in the reanalysis of the Match results. However, the Match results for that winter suggest that the ozone loss rates declined considerably by 31 March 1997 [*Schulz et al.*, 2000], so that the additional ozone loss in the last 12 days of the MLS study was probably small. The good agreement between the ozone losses in Table 1 can be reasonably expected to apply to the longer MLS period.

Match - vortex average ozonesondes

Estimates of the ozone loss in the 1996/97 winter have also been made using vortex averaged ozone mixing ratios from ozonesondes coupled with isentropic trajectories with adjustments for diabatic descent and mixing [*Knudsen et al.*, 1998]. The Match results have been reanalysed to cover the same time period. The same definition for the vortex edge could not be used as Knudsen used the maximum gradient in the PV fields while in Match the 36 s^{-1} isoline of nPV defines the vortex edge. However during the time period considered, the two definitions are found to be nearly identical. The results of the comparisons are given in Table 3 and Figure 2, and the agreement is generally good. The *Knudsen et al.* study reports ozone loss which is slightly, but not significantly, higher than the results from Match. These slight differences can be explained by differences in the

diabatic cooling rates used to adjust for vertical transport. The accumulated ozone losses that result from applying the diabatic rates used in Match to the Knudsen study are shown in parenthesis in Table 3, and there is better agreement with the Match results.

Lucic et al. [1999] developed a similar technique (but with vortex averaged ozone mixing ratios on isentropic surfaces) using ozonesonde measurements made in the 1991/92 winter. The vortex was stable for the first three weeks of January 1992 after which there was a major disturbance and in-flow of air from mid-latitudes. Their analysis of ozone loss was thus limited to the first three weeks of January during which they report an ozone loss of 0.32 ± 0.15 ppmv between 475 and 550K. When the same time period is considered, the ozone loss derived from Match is 0.3 ± 0.2 ppmv in good agreement with their results [*Rex et al.*, 1999]. The larger losses by Match for all of January [*von der Gathen et al.*, 1995] result from the significantly faster losses in the last part of January compared to the first part. The altitude distribution of the ozone loss found by the two approaches is also similar, with a small, statistically insignificant loss at 550K.

MLS - vortex average ozonesondes

The results from MLS [*Manney et al.*, 1997] and the vortex averaged ozone [*Knudsen et al.*, 1998] approaches have also been compared for 1996/97 and good agreement was found (Table 3 and Figure 2) despite the difference in the definition of the vortex edge.

Match – SAOZ/REPROBUS

Goutail et al. [1999] calculated the accumulated column ozone loss over the 1994/95 winter, using the 42 PVU isoline of potential vorticity at 475 K potential temperature as the definition of the vortex edge. This analysis has been repeated to match the time period and vortex definition of the Match study. For the period from 1 January to 31 March 1995, the total column loss derived from SAOZ is 117 DU while the partial column (380-600K) loss derived from Match is 127 ± 14 DU. The agreement is good, well within the estimated

uncertainty, and is consistent with the underlying assumption that the chemical ozone loss all occurred within the partial column covered by Match. Apart from 1994/95, the only winter where there is a sufficient number of individual matches to calculate column ozone losses from Match is 1999/2000 [*Rex et al.*, 2001]. Analysis shows similarly consistent results in that winter, with SAOZ/REPROBUS finding an accumulate loss of 105 DU for the whole column between 2 January and 25 March 2000, as against 88 ± 13 DU for the partial column found by Match (Figure 3 and Table 4). When POAM profile data for 1999/2000 are used in conjunction with the REPROBUS passive ozone instead of SAOZ data, a loss of 80 DU is found for the partial column 380-700 K over the same period.

Comparison of results from tracer correlation studies with explicit approaches

In this section results from work based on HALOE O_3/CH_4 and O_3/HF relations are compared with the results from the studies that account for dynamical processes by explicit transport calculations. It is hard to make the same detailed comparisons with the tracer correlation approach as the early winter measurements are so important in determining the total loss in the winter and the time evolution of the ozone loss is less well defined than for the other techniques. The problem is not the technique (see e.g., Richard et al [2001] for an analysis of the 1999/2000 winter using *in situ* data), but the limited (not continuous) sampling of HALOE measurements inside the vortex during the whole winter. However the good agreement between the various approaches which explicitly allow for transport allows us to concentrate on making comparisons between the total ozone losses derived from the tracer correlation and SAOZ approaches. The results of these comparisons are summarised in Table 4 for 1994/95, 1995/96, 1996/97 and 1999/2000 and are shown in Figure 3.

It is apparent that somewhat larger discrepancies exist between the results from the HALOE tracer correlation study and the results from SAOZ than between the results in the other comparisons. In 1994/95 and 1996/97 the tracer correlation approach found smaller

accumulated ozone losses than SAOZ. In 1995/96 the tracer study found larger losses than the SAOZ approach. Because there is good agreement between the SAOZ and Match approaches during the shorter period of comparison, there seems to be a larger uncertainty in the HALOE tracer correlation approach. This method to deduce ozone loss from tracer correlations is based on considering deviations from an early winter ozone-tracer reference relation. A proper reference relation must be measured late enough in the winter that a stable distinct vortex has already formed, and yet early enough that it is not already affected by chemical ozone loss.

The importance of the use of the proper reference relation is emphasised by the example of the winter 1996/97. In the original study *Müller et al.* [1997b] used a November HALOE reference; however, this reference is influenced by mixing in of ozone rich air into the polar vortex during late November/early December 1996. Moreover HALOE observations in December lie too far at the edge of the vortex. However it is possible to derive a representative reference function for early January 1997 from the ILAS satellite data set, which measured inside the vortex throughout the whole winter 1996/97. In early January 1997 the ozone-tracer relation is not changing inside the polar vortex [*Tilmes et al.*, Calculation of chemical ozone loss in the Arctic winter 1996/97 using ozone-tracer correlations: comparisons of ILAS and HALOE results, manuscript in preparation]. The more appropriate reference derived from the January ILAS data is characterised by larger ozone values and leads to the deduction of larger ozone losses (about 20%) in comparison to the use of the November relation in the study by Müller [1997b]. Using this reference, one obtains a vortex averaged column ozone loss of 64 ± 16 DU and an average loss in the vortex core (PV > 50 PVU at 475K) of 77 ± 17 DU, in better agreement with the loss derived from SAOZ/REPROBUS [*Goutail et al.*, 1998] (see Fig. 3 and Table 4).

Discussion

Detailed comparisons between the different studies that infer Arctic ozone loss from ozone

observations and transport calculations reveal a generally good agreement of the results. This degree of agreement is not obvious from the published data, since the time periods and regions considered differ widely. The agreement shows up only if the different approaches are applied for precisely the same times and regions. While there are similarities between the approaches that include explicit transport studies in that they use meteorological information, they use largely different types of instruments (ozonesondes, ground-based uv-visible spectrometers, satellite-borne microwave limb sounders and solar occulation instruments) to measure ozone and largely different approaches (trajectories of varying length and 3-D CTMs using different meteorological analyses) to separate the chemical loss from dynamical effects. However, with the exception of REPROBUS, the models used to calculate heating rates in these methods are similar, and in some cases the same. Diabatic heating rates in the lower stratosphere are difficult to determine accurately [Olaguer *et al.*, 1992] and more work on the validation of heating rates in different winters is required. In general, though, the good agreement between their results increases our confidence in our ability to quantify Arctic chemical ozone loss. At the same time, the importance of precise comparisons of ozone loss is clear, a point that needs to be borne in mind when comparing models with observations.

The studies relying on HALOE tracer relations to remove dynamical effects show somewhat larger discrepancies compared with the approaches that rely on transport calculations. These discrepancies vary from year to year. There are three possible reasons for this:

(a) quality of the HALOE measurements;

(b) uncertainty in the early winter relation, given HALOE's limited vortex sampling; and

(c) impact of mixing on tracer relations.

There is no evidence that the quality of the HALOE v.18 or v.19 data adversely affect the analyses of ozone loss. However it is important to use studies using these data rather than the earlier HALOE v.17 data.

In practice it is hard to separate the effects of (b) and (c). The comparisons presented here, along with the discussion in the original publications (particularly *Müller et al.* [2001]), indicate that a large uncertainty arises from the selection of the early vortex profiles used to determine the early winter ozone-tracer relations. With the ozone-tracer relation approach, the reference function should be calculated from profiles inside the early vortex, if the vortex is not influenced by significant mixing processes. If the baseline correlation function is derived too early, or around the edge of the vortex, it could have a significant impact on the overall derived ozone loss. For example, any mixing in December and early January could have a significant impact on the overall ozone loss derived from HALOE when the baseline correlations are derived from the late November measurements. Depending on the timing of the mixing relative to the ozone loss such mixing can lead to an overestimation or, more likely, an underestimation of the chemical loss. Great care, therefore, has to be taken to ensure that the baseline ozone-tracer relation is truly representative of the vortex prior to ozone loss in order for this approach to be valid. The influx of mid-latitude air into the Antarctic vortex during Austral winter [*Russell and Pierce*, 2000] would cause errors in ozone loss rates deduced there if the initial relation is taken inappropriately early.

A second possible reason for the larger discrepancies is that anomalous mixing in tracer space takes place during the periods of ozone loss. These considerations are in line with the arguments presented in *Michelsen et al.* [1998] and *Plumb et al.* [2000] which consider the effects of the mixing of mid-latitude air into the vortex. However the HALOE early winter relations are derived from measurements made inside the vortex that in general will lead to an underestimate of ozone loss in the vortex during winters when mixing is significant [*Müller et al.*, 2001]. Horizontal mixing is not significant inside an isolated vortex such as that in 1999/2000 [Ray *et al*., 2001]. The importance of its effect on the derived ozone loss will vary from year to year and will depend on the stability of the vortex during the periods of ozone loss. Given the preceding discussion about the importance of determining the

initial relation correctly, it seems likely that this mechanism (mixing during ozone loss) is best considered as contributing to the increased uncertainty associated with the ozone-tracer approach (rather than as the dominant cause).

A second sampling issue relates to the inhomogeneity of ozone loss inside the vortex, a point which is true for all methods used to derive vortex average ozone losses. The largest discrepancy (in 1996/97) is partially caused by the strong inhomogeneities inside the vortex that make it difficult to compare vortex averaged ozone losses [*Müller et al.*, 1997b; *McKenna et al.*, 2001; *Schulz et al.*, 2000]. In general though the careful comparisons presented here indicate that there is reasonably good agreement in the ozone losses derived from measurements using the techniques discussed.

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Figures

1. Ozone loss rates found by Match in the 1994/95 Arctic winter [*Rex et al.*, 1999]. The thick black lines show the subsiding air masses observed during the three periods for which an accumulated ozone loss is reported using MLS data [*Manney et al.*, 1996b]. The thick solid lines represent continuous periods of MLS observations, while the thick dashed line represents the air mass linking the final measurements in one north looking period of MLS with the early measurements in the next period. The solid blue line indicates the subsiding air mass closest to the air masses observed by MLS, and the dashed black lines indicate the paths of other subsiding air masses. In this study, the Match ozone loss rates are integrated along the thick black lines to allow a direct comparison with the MLS-derived losses. The isolines drawn with thin solid black lines show the area below PSC existence temperatures.
2. Comparison of accumulated losses in ozone mixing ratio derived from MLS [*Manney et al.*, 1996a,b, 1997], Match [*Rex et al.*, 1997, 1999; *Schulz et al.*, 2000] and the vortex average approach [*Knudsen et al.*, 1998] at potential temperatures around 475K. In each pair, the Match ozone loss rates have been integrated for the same time period and in the same subsiding air mass to allow direct comparisons.

3. Comparison of column ozone losses derived from the SAOZ/REPROBUS [*Goutail et al.*, 1998, 1999]; POAM/REPROBUS [*Deniel et al.*, 1998)]; Match [*Rex et al.*, 1999, 2001]; and HALOE tracer correlation [*Müller et al.*, 1996, 1997a, 1999; *Müller et al.*, Chemical ozone loss and chlorine activation deduced from HALOE and OMS measurements in the Arctic Winter 1999-2000, manuscript in preparation; *Tilmes et al.*, Calculation of chemical ozone loss in the Arctic winter 1996/97 using ozone-tracer correlations: comparisons of ILAS and HALOE results, manuscript in preparation] approaches. The 1996/97 January estimate uses ILAS data for the initial relation, and the 1999/2000 uses a mix of tracer data from the OMS remote and *in situ* balloon payloads which flew in November 1999. In both years the March relations used were found from HALOE data.

Tables

1) Ozone losses in the 1994/95 winter.
2) Ozone losses in the 1995/96 winter.
3) Ozone losses in the 1996/97 winter.
4) Column ozone losses.

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